



At sufficiently long times  $\tau$ , the coalescence rate  $\lambda_{i,j}(\tau)$  is expected to approach a finite limit  $\lambda_{i,j}^\infty$ . Successive collisions of a single particle then appear to be uncorrelated. They define a memoryless process and  $p_{i,j}(\tau)$  tends to the exponential distribution with rate parameter  $\lambda_{i,j}^\infty$ . The population-balance system (1)-(2) then reduces to

$$\dot{n}_i = \frac{1}{2} \sum_{j=1}^{i-1} \lambda_{i-j,j}^\infty n_{i-j} n_j - \sum_{j=1}^{\infty} \lambda_{i,j}^\infty n_i n_j. \quad (4)$$

This is the celebrated Smoluchowski coagulation equation [20]. The stationary rates  $\lambda_{i,j}^\infty$  are usually referred to as the collection or coalescence kernels. The work cited above on particle inertia was actually devoted to estimating their dependence upon particle sizes and the turbulent fluctuations of the carrier flow. The kinetic model (4) leads to predictions concerning the short-time increase of the  $n_i$ 's. At the early stages of particle growth, the number  $n_1$  of monomers remains almost constant and creations are dominant in the population balance. We thus get  $\dot{n}_2 \simeq \lambda_{1,1}^\infty n_1^2/2$ , so that  $n_2(t) \simeq n_1^2 \lambda_{1,1}^\infty t/2$ . For the next size, we have  $\dot{n}_3 \simeq \lambda_{1,2}^\infty n_1 n_2$  and thus  $n_3(t) \simeq n_1^3 \lambda_{1,1}^\infty \lambda_{1,2}^\infty t^2/4$ . We obtain recursively

$$n_i(t) \simeq n_1^i (t/t_i)^{i-1}, \quad (5)$$

where the times  $t_i$  are averages of the times  $1/\lambda_{j,k}^\infty$  associated to the different combinations of coalescences  $(j)+(k)$  that are necessary to form a particle  $(i)$ . The consistency of the assumptions can be checked a posteriori: The creation terms in (4) are always  $\propto t^{i-2}$  and thus prevail at short times over the dominant destruction term  $\propto t^{i-1}$ .

The main assumption leading to Smoluchowski kinetics (4) is a convergence of the coalescence rate to its limiting value  $\lambda_{i,j}^\infty$  much faster than the evolution of  $n_i$ . This is ensured for instance when the particles are very dense. For explaining the formation of large particles in a dilute suspension, these timescales are in general not sufficiently separated. The sudden appearance of sizable aggregates requires a brisk sequence of coalescences that are very likely to be correlated to each other. When, in addition, the coalescing species are transported by a turbulent flow, such correlations speed up the growth of large particles.

A statistically steady turbulent flow involves interactions between eddies of various sizes, ranging from the integral scale  $L$ , where kinetic energy is injected at a rate  $\varepsilon$ , down to the dissipative scale  $\eta = \nu^{3/4}/\varepsilon^{1/4}$  below which viscous damping dominates ( $\nu$  denotes the kinematic viscosity of the fluid). The degree of turbulence grows with the extension of this spatial span and is measured by the Reynolds number  $Re = (L/\eta)^{3/4}$ . The intermediate scales between  $\eta$  and  $L$  define the inertial range through which energy cascades with a rate  $\varepsilon$ . Dimensional arguments suggest that the velocity increments between two points separated by a distance  $r$  in the inertial range behave as  $u_r \sim (\varepsilon r)^{1/3}$ . Such a phenomenology, referred to

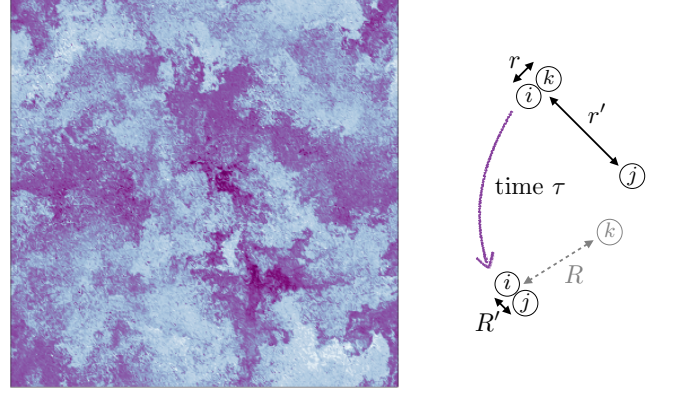


FIG. 1. *Left*: Distance traveled by fluid elements in a 3D turbulent flow during one large-eddy turnover time. Long (white) and short (purple) distances, represented here as a function of the final position in a 2D slice, define an intricate landscape with fronts where particles coming from far apart meet together. *Right*: Sketch of the event leading to correlated successive collisions. At the initial time (top), two particles  $i$  and  $k$  are located at a collision distance  $r \lesssim \eta$ , while a third one  $j$  is at distance  $r' \gg \eta$  far from them. A time  $\tau$  later (bottom)  $j$  has approached  $i$  at a distance  $R' \lesssim \eta$  while  $k$ , having collided or not, has escaped to  $R \gg \eta$ .

as Kolmogorov 1941, is often enough for capturing the most significant effects of turbulent fluctuations; in reality the scaling properties display slight deviations, due to intermittency, from this dimensional prediction [21, 22].

The breakdown of scale invariance is much more striking for mixing statistics, owing to the fact that turbulence mingles together fluid elements in a robust manner. This pops up with the presence of quasi-discontinuities in the Lagrangian map where materials originating from distinct regions of the flow are violently brought together (see Fig. 1 *Left*). The emergence of such *fronts* is due to the inertial-range roughness of the velocity field and the associated non-uniqueness of fluid element trajectories. Two initially separate tracers  $\mathbf{x}_1(t)$  and  $\mathbf{x}_2(t)$  that closely approach each other become indistinguishable and separate afterwards following Richardson's superdiffusion  $|\mathbf{x}_1(t) - \mathbf{x}_2(t)|^2 \sim \varepsilon t^3$ . Still, when interested in more than two fluid elements, this explosive behavior is constrained by the underlying presence of *statistical conservation laws* induced by the spatial correlations of the velocity field [18, 19, 23]. There exists specific functions of the shape and size of a cloud of  $n$  tracers that on average do not vary with time. Such zero modes are known to yield anomalous scaling in the statistics of an advected passive scalar  $\theta$ . Its structure functions behave in the inertial range as  $\langle (\theta(\mathbf{x} + \mathbf{r}) - \theta(\mathbf{x}))^n \rangle \sim |\mathbf{r}|^{n/3 - \delta_n}$  where the discrepancies  $\delta_n$  of the exponents from their dimensional prediction relate to the anomalous scaling of the transition probability of the distances between  $n$  tracers. As we will now see, the behavior of the three-point motion is in fact of relevance to coalescences.

In dilute suspensions, a coalescence results from two

successive processes. First, the turbulent flow needs to bring two initially separate particles at a sufficiently close distance  $\lesssim \eta$ . Second, these close particles need to actually merge, and this involves various microphysical mechanisms (particle inertia, hydrodynamical interactions, surface effects). This leads us to write the coalescence rate as a product of two contributions:

$$\lambda_{i,j}(\tau) \approx \lambda_{i,j}^{\text{turb}}(\tau) \times \lambda_{i,j}^{\text{micro}}. \quad (6)$$

The contribution from turbulent transport can be written

$$\lambda_{i,j}^{\text{turb}}(\tau) \approx \int u_\eta p_3(R, \eta, \tau | \eta, r', 0) (r'^2/L^3) dr' dR \quad (7)$$

involving the transition probability  $p_3$  of the three-point motion. More specifically, two successive collisions occur if three particles (see Fig. 1 *Right*), initially separated by distances  $r = \eta$  and an arbitrary  $r'$ , come in a time  $\tau$  to distances  $R$ , arbitrary, and  $R' = \eta$ . The relation (7) is obtained by integrating over all possible initial distances  $r'$  of the particle  $\textcircled{j}$  from the particle  $\textcircled{i}$ , with a weight  $\propto r'^2$  given by a uniform three-dimensional spatial distribution. The three tracers  $\textcircled{i}$ ,  $\textcircled{j}$  and  $\textcircled{k}$  undergo in a time  $\tau$  an evolution from a degenerate triangle with  $r \ll r'$  to another degenerate triangle with, this time,  $R \gg R'$ . In turbulence, the probability transition between such configurations can be written as

$$p_3(R, R', \tau | r, r', 0) \approx \left(\frac{\eta}{r'}\right)^2 \left(\frac{L}{r'}\right)^{\delta_3} \frac{1}{\varepsilon \tau^3} \Psi\left(\frac{r'^2}{\varepsilon \tau^3}, \frac{R^2}{\varepsilon \tau^3}\right),$$

where  $\delta_3$  is the anomalous part of the scaling exponent associated to the third-order statistics of an advected passive scalar; its value is universal (independent of the injection mechanism) and  $\approx 0.18$ , as reported from several experimental and numerical studies [24]. In the expression above the first factor comes from integration over angles and can be seen as a small solid-angle contribution. The second factor originates from intermittency and gives a dependence upon the integral scale  $L$ . Physically, it means that when  $\delta_3 > 0$ , the closer is the third particle, the more likely it is to approach one of the other two. The last terms involve a dimensionless function  $\Psi$  that imposes Richardson's scaling for backward and forward pair evolution. This specific form of the three-point transition probability leads to

$$\begin{aligned} \lambda_{i,j}^{\text{turb}}(\tau) &\propto \frac{\nu^{7/4}}{\varepsilon^{1/4} L^{3-\delta_3}} \int \Psi\left(\frac{r'^2}{\varepsilon \tau^3}, \frac{R^2}{\varepsilon \tau^3}\right) \frac{dr' dR}{r'^{\delta_3} \varepsilon \tau^3} \\ &\propto (1/\tau_L) (\tau/\tau_L)^{-\frac{3}{2}\delta_3}, \end{aligned} \quad (8)$$

where  $\tau_L = \varepsilon^{-1/3} L^{2/3}$  is the large-eddy turnover time. Turbulent transport thus leads to a power-law dependence in time of the coalescence rate. Plugging this behavior in the global coalescence rate (2) and by using the

population-balance equations (1), one obtains a short-time behavior of the number of particles  $\textcircled{i}$  that reads

$$n_i(t) \simeq n_1^i (t/\tilde{t}_i)^{(1-\frac{3}{2}\delta_3)(i-2)+1}. \quad (9)$$

Here the characteristic times  $\tilde{t}_i$  are  $\propto i \tau_L$ , with a proportionality constant that involves the various microphysical rates  $\lambda_{j,k}^{\text{micro}}$  of the coalescences leading to  $\textcircled{i}$ . For  $\delta_3 > 0$  the algebraic exponent appearing in (9) is smaller than that obtained in (5) from Smoluchowski's kinetics. The intermittency of turbulence mixing is thus enhancing the short-time growth by coalescence. In addition, the larger is the aggregate size considered, the stronger is this enhancement. Indeed, when  $i$  is large and  $t \ll i \tau_L$ , the formation of a particle  $\textcircled{i}$  requires a large number of correlated coalescences separated by inertial-range times and the population dynamics is dominated by (9).

In order to corroborate our theoretical predictions on the enhancement of coalescences by turbulent mixing, we have performed direct numerical simulations for the evolution of a dilute population suspended in a turbulent flow. We start from one billion inertial point-particles whose dynamics is given by a viscous Stokes drag:

$$\ddot{\mathbf{x}}_n = -\frac{1}{\tau_n} [\dot{\mathbf{x}}_n - \mathbf{u}(\mathbf{x}_n, t)], \quad (10)$$

where  $\mathbf{u}$  designates the fluid velocity field. It is obtained numerically by a pseudo-spectral integration of the incompressible Navier-Stokes equation using  $2048^3$  gridpoints. A large-scale forcing is applied in order to maintain the flow in a developed turbulent state with  $Re \approx 50\,000$ .

The particles follow the flow with a time lag given by their individual response times  $\tau_n$ . Each particle has a virtual radius  $a_n(t)$  and  $\tau_n \propto a_n^2$ . We start from a monodisperse suspension with monomers having initially all the same radius  $a_i(0) = a_0 \approx \eta/10$ . When two particles approach at a distance equal to the sum of their radii (detected using a billiard algorithm), they merge, conserving mass and momentum. Particles inertia is measured by their Stokes numbers  $St_n = \tau_n \varepsilon^{1/2} / \nu^{1/2}$ , which is initially small  $St_n(0) \approx 0.1$ . Inertia effects can thus be clearly neglected when interested in inertial-range length or time scales. The suspension is dilute: their volume fraction is approximately  $5 \times 10^{-5}$ , which represents in our flow one particle for each cube of volume  $10 \eta^3$  and is consistent with, for example, typical settings in a warm cloud of our atmosphere.

Figure 2 shows on log-log scales the time evolution of the number  $n_i(t)$  of particles made from the merger of  $i$  monomers (that is with radius  $a(t) = i^{1/3} a_0$ ) for  $i = 1, 2, \dots, 15$ . Data (dots) is approximated very well by  $n_i(t) \propto t^{0.73(i-2)+1}$  (dashed lines), corresponding to the predicted power laws (9) with  $\delta_3 = 0.18$ . Such a behavior persists for times larger than the large-eddy turnover

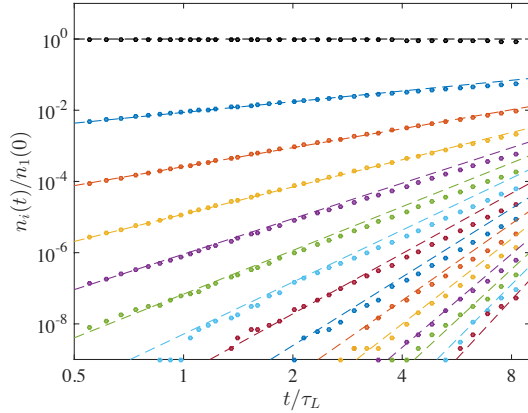


FIG. 2. Time evolution of the number  $n_i(t)$  of particles (normalized by the initial number of monomers  $n_1(0)$ ); the mass  $i$  increases from 1 to 15 from top to bottom. The dots are the results of direct numerical simulations and the dashed lines show for  $i \geq 2$  behaviors  $\propto t^{0.73(i-2)+1}$  deduced from (9) for  $\delta_3 = 0.18$ .

time. The result of our simulation confirms the accuracy and the relevance of the predictions made earlier in this Letter. Large aggregates are appearing faster than predicted from kinetic models.

To confirm that this enhanced growth is indeed resulting from correlated successive collisions, we have measured the probability density  $p_{i,1}(\tau)$  of the time lag  $\tau$  between the creation of a particle ( $i$ ) and its next collision with a ①. Results are shown in Fig. 3 for  $i = 2, 3, 4, 5$ . The distributions clearly display for  $\tau$  in the inertial range a power-law decay, followed by a (stretched) exponential cutoff at  $\tau \gtrsim \tau_L$ . The measured value of the algebraic exponent is consistent with the predicted value  $-(3/2)\delta_3 \approx -0.27$ . This confirms that the inter-collision time distribution follows (3) with a time-dependent coalescence rate  $\lambda_{i,j}(\tau) \propto \tau^{-\frac{3}{2}\delta_3}$ .

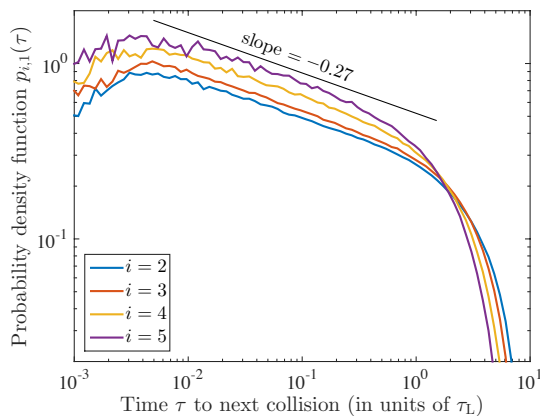


FIG. 3. Probability density function of the time between successive coalescences of a particle  $i$  with a monomer of mass 1. The power-law tail has, as we predict theoretically, an exponent  $\approx -0.27 \approx -(3/2)\delta_3$

In conclusion, let us stress again that intermittency of turbulent mixing is responsible for an enhanced growth of dilute coalescing aggregates. To our knowledge, this is one of the first instances where turbulent anomalous scaling laws play a critical role in describing to leading order a process with practical implications. Here, only third-order statistics are relevant since successive binary collisions involves the evolution of triplets of tracers that form degenerate triangles. Higher-order statistics enter other configurations (when, for instance, two particles are simultaneously formed and then merge) but they give subleading contributions. Finally, it is worth mentioning that the effect unveiled here might be accounted for by modifying kinetic models. When coarse-graining the population dynamics on sufficiently large timescales, correlated successive collisions will then appear as simultaneous multiple collisions.

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- [1] S. Weidenschilling and J. N. Cuzzi, in *Protostars and Planets III*, Vol. 1 (1993) pp. 1031–1060.
  - [2] J. J. Lissauer, *Ann. Rev. Astron. Astrophys.* **31**, 129 (1993).
  - [3] M. Pinsky and A. Khain, *J. Aerosol Sci.* **28**, 1177 (1997).
  - [4] R. Shaw, *Annu. Rev. Fluid Mech.* **35**, 183 (2003).
  - [5] G. Wetherill and G. R. Stewart, *Icarus* **77**, 330 (1989).
  - [6] A. B. Kostinski and R. A. Shaw, *Bull. Am. Met. Soc.* **86**, 235 (2005).
  - [7] A. Johansen, M.-M. Mac Low, P. Lacerda, and M. Bizzarro, *Science Advances* **1**, e1500109 (2015).
  - [8] A. Johansen *et al.*, *Nature* **448**, 1022 (2007).
  - [9] E. Bodenschatz, S. Malinowski, R. Shaw, and F. Stratmann, *Science* **327**, 970 (2010).
  - [10] S. Sundaram and L. Collins, *J. Fluid Mech.* **335**, 75 (1997).
  - [11] G. Falkovich, A. Fouxon, and M. G. Stepanov, *Nature* **419**, 151 (2002).
  - [12] M. Wilkinson, B. Mehlig, and V. Bezuglyy, *Phys. Rev. Lett.* **97**, 048501 (2006).
  - [13] B. Devenish *et al.*, *Q.J.R. Meteorol. Soc.* **138**, 1401 (2012).
  - [14] L. Pan and P. Padoan, *Astrophys. J.* **797**, 101 (2014).
  - [15] J. Bec, A. Celani, M. Cencini, and S. Musacchio, *Phys. Fluids* **17**, 073301 (2005).
  - [16] K. Gustavsson and B. Mehlig, *Phys. Rev. E* **87**, 023016 (2013).

- (2013).
- [17] M. Voßkuhle, A. Pumir, E. Lévêque, and M. Wilkinson, *J. Fluid Mech.* **749**, 841 (2014).
  - [18] B. I. Shraiman and E. D. Siggia, *Nature* **405**, 639 (2000).
  - [19] G. Falkovich, K. Gawędzki, and M. Vergassola, *Rev. Mod. Phys.* **73**, 913 (2001).
  - [20] M. Smoluchowski, *Zeitschrift f. physik. Chemie* **92**, 129 (1917).
  - [21] U. Frisch, *Turbulence* (Cambridge University Press, Cambridge, England, 1995).
  - [22] G. Falkovich and K. R. Sreenivasan, *Physics Today* **59**, 43 (2006).
  - [23] I. Arad, L. Biferale, A. Celani, I. Procaccia, and M. Vergassola, *Phys. Rev. Lett.* **87**, 164502 (2001).
  - [24] Z. Warhaft, *Ann. Rev. Fluid Mech.* **32**, 203 (2000).